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(54) Process for the Production of  
 Anti-Static Biaxially Oriented  
 Polypropylene Composite Films and  
 Product of the Process

(57) A polypropylene substrate layer is  
 formed from a polypropylene resin  
 comprising 100 parts by weight of  
 polypropylene and from about 0.3 to  
 about 1 part by weight of at least one  
 anti-static agent selected from the  
 fatty acid glycerides of the following  
 formula:



wherein R is a fatty acid residue

(monovalent aliphatic hydrocarbon  
 group) and coated on one surface or  
 both surfaces with a polyolefin layer to  
 form a two- or three-layered integral  
 composite film. The polyolefin is a  
 polyethylene or an ethylene-( $\alpha$ -olefin)  
 copolymer. The polypropylene  
 substrate film is stretching in a first  
 direction to orient the film uniaxially,  
 and then one surface or both surfaces  
 is covered with the polyolefin layer in  
 a manner which forms a laminate. The  
 laminated film is then stretched in the  
 direction transverse to the first  
 direction to form the biaxially oriented  
 composite film.

ANTI-STATIC BIAXIALLY ORIENTED POLYPROPYLENE  
 COMPOSITE FILM + CONTAINS FATTY ACID  
 MONOGLYCERIDE IN POLYPROPYLENE LAMINATED  
 TO ETHYLENE POLYMER

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## SPECIFICATION

## Process for the Production of Anti-Static Biaxially Oriented Polypropylene Composite Films and Product of the Process

5 The present invention relates to a process for the production of anti-static biaxially oriented polypropylene composite films and the products of said process. More particularly, the invention relates to a process for ready and economical production of biaxially oriented transparent polypropylene composite films having excellent anti-static properties which are readily sealable and have satisfactory mechanical strength. 5

10 Biaxially oriented polypropylene films are widely used as wrapping material, particularly for food, because they are transparent, glossy, odorless and non-toxic and have extremely low vapor permeability and a useful level of mechanical strength. These polypropylene films, however, have an inherent defect in that, due to their hydrophobic nature, they readily accumulate a static charge, which is one of the characteristic properties common to all resins. Such static charge generally brings about various disadvantages such as, for example, that dust is attracted to and collects on the surface of the film, thus spoiling the appearance of the merchandise; the charged surface has poor printability; in the case of bag making, the bags do not stack evenly; and operators sometimes get an electric shock from the discharge of the accumulated electrostatic charge, and so forth. 15

20 To prevent such difficulties and disadvantages caused by static charge, anti-static agents are generally applied to the biaxially oriented polypropylene films. An anti-static agent may be applied by coating it on the surface of the film. Alternatively, an anti-static agent may be incorporated as an internal additive into the polypropylene composition before it is formed into a film to be subjected to a biaxial orientation treatment. 20

25 Film coated with an anti-static agent suffers from a number of disadvantages. When the anti-static coating layer is removed from the surface of the film in handling, etc., the same surface is no longer anti-static. Such coatings generally decrease the transparency and the anti-blocking properties of the film. The cost of production of anti-static coated films generally increases because it requires an additional process step for the application of the coating formed from the anti-static agent after the production of the biaxially oriented films. 25

30 When the anti-static agents are used by incorporating them into the polypropylene composition before the biaxially oriented film is made therefrom, the resulting film products will be free from the disadvantages of the coating method. However, up to now, incorporating the anti-static agent in the composition has been effective only when (i) special types of expensive compounds are used as the internal additive; and/or (ii) the propylene composition is selected from very specific blends of propylene with specified copolymers; and in addition the resulting films require corona discharge treatment. Representative compounds of the prior art useful as an internal additive anti-static agent include amphoteric surface active agents such as, for example, the adducts of alkyl amine with ethylene oxide, the imidazolines, the betaines and the like. All of these compounds are very expensive compared with the fatty acid monoglycerides which can be used in the coating method. 35

40 Japanese Patent Publication No. 33827/75 (published on November 4, 1975) discloses that a mixture of a fatty acid monoglyceride and at least 50% by weight of a betain-type amphoteric surface active agent can be used as an internal additive for improving anti-static properties of oriented polypropylene films. In addition, it is essential to subject the finished film products to corona discharge treatment after the biaxial orientating. 40

45 Japanese Patent Publication No. 792/78 (published on January 12, 1978) discloses that a fatty acid monoglyceride is effective as an internal type anti-static agent when incorporated into a polymer blend of polypropylene and ethylene-vinyl acetate copolymer or ethylene-acrylic ester copolymer, though the same is not effective when used incorporated in polypropylene alone. In this instance, also, it is essential to treat the polymer blend film by corona discharge after the biaxial orientation thereof to obtain the desired anti-static properties. 45

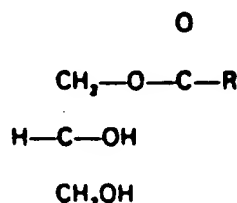
50 Heretofore, the inexpensive fatty acid monoglycerides could not be used effectively as the internal anti-static agent to be incorporated in an essentially all polypropylene composition to be used in preparing a biaxially oriented polypropylene film, because the anti-static properties would be lost 50

55 It is an object of the present invention to provide anti-static biaxially oriented polypropylene composite films useful as wrapping material prepared using inexpensive fatty acid monoglycerides as the effective internal additive. 55

60 The present invention provides biaxially oriented polypropylene composite films having good transparency and satisfactory anti-static properties utilizing as the internal additive one or more inexpensive fatty acid monoglycerides for the purpose of improving anti-static properties of the resulting biaxially oriented polypropylene composite films. 60

According to the present invention, anti-static biaxially oriented polypropylene composite films are produced by forming a film, preferably by melt extrusion, from a composition comprising 100 parts by weight of polypropylene and from 0.3 to 1.0 part by weight of at least one anti-static agent selected from the fatty acid monoglycerides having the generic formula:

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wherein R represents a monovalent aliphatic hydrocarbon group which is a fatty acid residue; stretching the resulting film in one direction, e.g., the longitudinal direction, to obtain a uniaxially oriented film; forming on one or both surfaces of said uniaxially oriented film, a layer of polyethylene and/or an ethylene-( $\alpha$ -olefin) copolymer to obtain a laminated film; and then stretching the laminated film in the transverse direction to obtain a final product of an anti-static, biaxially oriented polypropylene composite film. The monovalent aliphatic hydrocarbon group R is preferably selected from those which have from 9 to 17 carbon atoms in their hydrocarbon chains. The invention also provides the novel two-ply and three-ply laminates produced by the aforescribed process, and exemplified hereinafter.

The polypropylene composition used to form the polypropylene film may be formed from propylene and/or copolymers of propylene with one or more monomers so long as the copolymers have an extraction residue determined in boiled heptane of 85% by weight or higher and at least 90% by weight of the copolymer component is polypropylene. The polypropylene composition may contain, as occasion demands, known propylene adjuvants such as, an anti-oxidant, an ultraviolet UV absorber, an anti-blocking agent, a lubricant and the like, in addition to said inexpensive anti-static agent.

The fatty acid monoglycerides in the polypropylene composition which are useful as the internal anti-static agents include mixtures of mono-, di- and tri-glycerides so long as the mono-glyceride component forms at least about 90% of each of said mixtures. That is, the mixture can contain up to about 10% of di- and tri-glyceride components. From 0.3 to 1.0 part by weight of such fatty acid monoglyceride is incorporated in 100 parts by weight of the polypropylene composition. If the amount of said additive is less than the minimum specified above, satisfactory anti-static properties will not be obtained, and if the addition is excessive, the transparency generally required for packing films will not be obtained.

The polypropylene composition is preferably formed by melt extrusion into a film which is then uniaxially oriented, and a polyethylene or an ethylene-( $\alpha$ -olefin) copolymer resin is laminated on the resulting uniaxially oriented film. The ethylene-( $\alpha$ -olefin) copolymer resins contain units of "ethylene" and at least one other " $\alpha$ -olefin". Representative examples for useful ethylene-( $\alpha$ -olefin) copolymer resins include ethylene-propylene copolymer; ethylene-butene copolymer; ethylene-propylene-butene terpolymer; and mixtures of these. These copolymer resins may contain, as occasion demands, polyolefin adjuvants, such as, an anti-oxidant, a UV absorber, an anti-blocking agent, a lubricant and the like.

The laminated film of the present invention comprising the biaxially stretched polypropylene film having laminated thereon a layer of ethylene and/or the ethylene-( $\alpha$ -olefin) copolymer resin can be prepared either by extrusion laminating the polyethylene or ethylene-( $\alpha$ -olefin) copolymer resin on the uniaxially oriented polypropylene film or alternatively by adhering, preferably by pressure adhering, a preformed film of the polyethylene or ethylene-( $\alpha$ -olefin) copolymer resin on the uniaxially oriented polypropylene film.

The anti-static properties of the surface of the laminated polyethylene or ethylene-( $\alpha$ -olefin) copolymer resin layer of the biaxially oriented polypropylene composite film prepared by the process of the present invention can be further improved, for example, by subjecting the composite film to corona discharge treatment.

When treating a two-ply laminated film (a film having a lamination layer on only one surface of a substrate polypropylene layer), it is preferred that only the biaxially oriented polypropylene resin layer should be exposed to the corona discharge, in order not to impair the heat sealability of the laminated polyethylene or ethylene-( $\alpha$ -olefin) copolymer resin layer. This is also preferred since print or other surface decoration is generally applied to the surface opposite the surface having the good heat sealability.

The corona discharge treatment of the outer (exposed) surface of the polypropylene substrate layer of the biaxially oriented laminated film causes a remarkable improvement in anti-static properties of the surface of the polyethylene or ethylene-( $\alpha$ -olefin) copolymer resin layer laminated on said substrate layer of the laminated film. When each side (or surface) of a polypropylene substrate layer has a laminated polyethylene or ethylene-( $\alpha$ -olefin) copolymer lamination outer layer thereon, either one of these outer surface layers can be treated by corona discharge.

The process of the present invention provides for economical and ready production of transparent, biaxially oriented polypropylene composite films which have excellent anti-static properties and have good heat-sealability and satisfactory mechanical strength by making use of the

conventional tentering-type (stretching) sequential biaxially orienting apparatus without requiring modification of its operation.

The two-layer anti-static biaxially oriented composite films are generally in a total thickness between about 10 and 100 microns, and more preferably between about 15 and 60 microns. The one laminated polyethylene and/or ethylene-( $\alpha$ -olefin) copolymer layer is preferably between about 0.5 and 10 microns, and more preferably between about 1.0 and 6.0 microns. There is no specific lamination with respect to the ratio of the thickness of the polypropylene substrate layer to the lamination layer so long as the above mentioned requirements are fulfilled. However, particularly in the case of very thin composite films, the polypropylene substrate layer should occupy at least 50% of the total thickness of the composite film.

Similarly, the three-layer anti-static biaxially oriented composite films are generally in a total thickness between about 10 and 100 microns, and more preferably between about 15 and 60 microns. Each of the two laminated polyethylene and/or ethylene-( $\alpha$ -olefin) copolymer layers is preferably between about 0.5 and 10 microns, and more preferably between about 0.8 and 4.0 microns.

As regards the ratio of the thickness of the polypropylene substrate layer to each of the two lamination layers, the same is true as mentioned above with respect to the two-layer anti-static biaxially oriented composite films.

The biaxially oriented polypropylene composite films obtained according to the process of the present invention exhibit, as they are, satisfactory anti-static properties as well as good lubricity and anti-blocking properties, and accordingly, they can be used as packaging material to be used in automatic form-and-fill packaging machines. However, if corona discharge treatment is also employed, biaxially oriented polypropylene composite films having greatly improved characteristic properties are obtained.

The following examples will illustrate the process of the present invention more concretely and also demonstrate the improved characteristic properties of the biaxially oriented polypropylene composite films of the present invention.

#### Example 1

Seven compositions were prepared. Each composition, which contained 100 parts by weight of polypropylene having an  $[\eta]$  determined in tetralin at 135°C. of 1.9 and an n-hexane insoluble component of 90% or higher, 0.1 part by weight of stearic amide, and 0.5 part by weight of one of the fatty acid monoglycerides set forth in Table 1, was melt extruded at an extrusion temperature of 280°C. to form a film which was then oriented in the machine direction (longitudinally) and roller stretched to five times its original length. The surface of the roller was heated to a temperature of 140°C. to obtain a uniaxially oriented polypropylene film 160 microns thick.

Onto the resulting uniaxially oriented polypropylene film was extrusion laminated an ethylene-propylene copolymer comprising 4% by weight of ethylene and 96% by weight of propylene and having a melting point of 140°C. and an M.I. (melt index) of 7.0 to form thereon a lamination layer 40 microns ( $\mu$ ) thick to obtain a laminated film 200 microns thick. The resulting laminated film was then transversely oriented by stretching its width by a multiple of eight times at an orientation temperature of 160°C. to obtain a biaxially oriented polypropylene composite film 25 microns thick. All of the above-mentioned steps were carried out in one continuous process.

After 48 hours of aging in the atmosphere at a temperature of 40°C., the resulting composite film prepared in each run set forth in Table 1 was tested to determine its characteristic properties, such as anti-static properties, anti-blocking properties, transparency, lubricity and the like. The results are shown in Table 1.

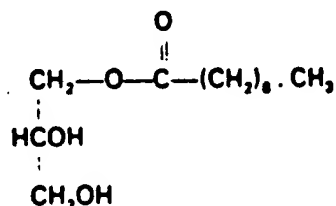
For comparative purposes, a biaxially oriented polypropylene composite film was prepared (Run No. 8) in the same manner as in Example 1 except that 0.5 part by weight of a betain-type anti-static agent, stearyl-di- $\beta$ -hydroxyethyl-betain, instead of a fatty acid monoglyceride, was incorporated in 100 parts by weight of polypropylene. It was also tested and the results reported in Table 1, for comparative purposes.

Table 1

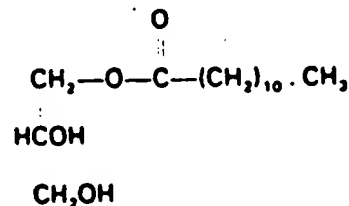
			Ratio of electro- static decay	Surface resis- tivity ( $\Omega/\text{cm.}$ )	Anti- blocking proper- ties (class)	Coef- ficient of friction	
5	Run No.	Anti-static agent	Haze (%)	(%)			5
	1	decanoyl monoglyceride	2.8	20	$1 \times 10^{13}$	3—4	0.50
	2	lauryl monoglyceride	2.8	20	$1 \times 10^{13}$	2—3	0.45
	3	myristyl monoglyceride	2.8	20	$1 \times 10^{13}$	1—2	0.35
	4	palmityl monoglyceride	2.6	30	$7 \times 10^{12}$	1	0.25
10	5	stearyl monoglyceride	2.7	60	$2 \times 10^{12}$	1	0.25
	6	a mixture of palmityl monoglyceride (70) and stearyl monoglyceride (30)	2.5	40	$8 \times 10^{12}$	1	0.25
	7	a mixture of palmityl monoglyceride (30) and stearyl monoglyceride (70)	2.6	50	$4 \times 10^{12}$	1	0.25
15	8	stearyl di- $\beta$ - hydroxyethyl-betain	2.4	0	$1 \times 10^{16}$ or higher	1	0.25
							15

The structural formula of each of the compounds used as anti-static agents in the runs in Table 1 follows:

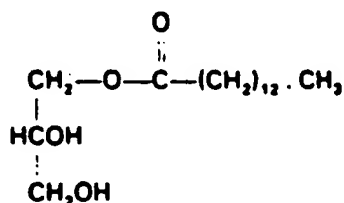
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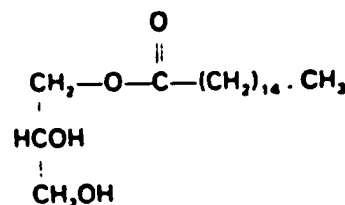
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No. 3

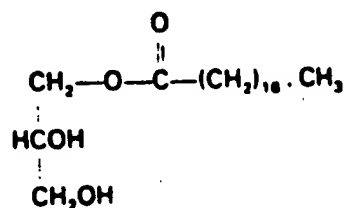


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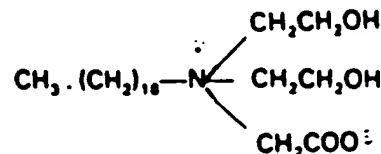


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No. 5



No. 8



25

## Example 2

The biaxially oriented polypropylene composite films, 25 microns thick, obtained in Runs 1 through 7 set forth in Table 1 were subjected to corona discharge at the outer surface of the polypropylene film side of the composite film and then the thus-treated composite film was aged in the atmosphere at a temperature of 40°C. for 48 hours. Then, the characteristic properties, including anti-static properties, anti-blocking properties, transparency, lubricity and the like, were determined as recited in Example 1. The results are shown in Table 2 below.

30

Table 2  
Ratio of  
electro-  
static  
decay

Run No.	Anti-static agent	Haze (%)	Ratio of electrostatic decay (%)	Surface resistivity ( $\Omega/\text{cm.}$ )	Anti-blocking properties (class)	Coefficient of friction
9	decanoyl monoglyceride	2.9	30	$7 \times 10^{12}$	3—4	0.45
10	lauryl monoglyceride	2.9	30	$7 \times 10^{12}$	2—3	0.43
11	myristyl monoglyceride	3.0	40	$6 \times 10^{12}$	1—2	0.33
12	palmityl monoglyceride	2.8	50	$4 \times 10^{12}$	1	0.25
13	stearyl monoglyceride	3.0	90	$3 \times 10^{11}$	1	0.25
14	a mixture of palmityl monoglyceride (70) and stearyl monoglyceride (30)	2.7	60	$2 \times 10^{12}$	1	0.25
15	a mixture of palmityl monoglyceride (30) and stearyl monoglyceride (70)	2.8	80	$1 \times 10^{12}$	1	0.25

Both the "ratio of electrostatic decay" and the "surface resistivity" were determined with respect to the outer surface of the lamination (copolymer) layer side of each of the tested composite films.

Both the "anti-blocking properties" and the "coefficient of friction" were determined with respect to the two contacting surfaces: the outer surface of the lamination layer side of a test strip of a composite film against the outer surface of the same side of another test strip of the same composite film.

### Example 3

Uniaxially oriented polypropylene films were prepared in the same manner as in Example 1 using a mixture of palmityl monoglyceride and stearyl monoglyceride in a ratio of 30:70 by weight as the anti-static agent in the amounts as set forth in Table 3, incorporated in 100 parts by weight of polypropylene pellets of the same type used in Example 1 which contained 0.1 part by weight of stearic amide. Then, each of the ethylene-( $\alpha$ -olefin) resins listed in Table 3 was extrusion laminated onto one surface of each of said uniaxially oriented polypropylene films and then oriented in the same manner as in Example 1 to obtain the desired biaxially oriented polypropylene composite film 25 microns thick in one continuous process.

The composite films thus prepared were aged in the atmosphere at a temperature of 40°C. for 48 hours and then tested as in Example 1 to determine their characteristic properties, such as anti-static properties, anti-blocking properties, transparency and lubricity. The surface of the biaxially oriented polypropylene film layer of these biaxially oriented composite films was then subjected to corona discharge treatment and aged in the atmosphere at a temperature of 40°C. for 48 hours. Then, the same tests with respect to the above-mentioned characteristic properties were repeated. The results of these Runs 16—27 are also reported in Table 3.

For comparative purposes, biaxially oriented polypropylene composite films were prepared in the same manner as above according to the process of the present invention except that the fatty acid monoglyceride was incorporated in the polypropylene composition in an amount outside the range defined in the claims with respect to the fatty acid monoglyceride. The characteristic properties such as anti-static properties of these films were also determined in the same manner as hereinbefore mentioned and results are also shown in Table 3.

The ethylene-( $\alpha$ -olefin) resins used in these runs had the following properties:

low density polyethylene: MI: 7.0, density: 0.920;

high density polyethylene: MI: 4.0, density: 0.950;

ethylene-butene copolymer: MI: 5.0, density: 0.900;

butene content: 10 mole %;

ethylene-butene-propylene terpolymer:

$[\eta]$  (viscosity): 1.78,

butene content: 5.0 weight %,

ethylene content: 0.6 weight %.

Table 3

		Amount of anti- static agent added (part by wt.)	Type of polymer used as lamination layer	Corona discharge treatment	Haze (%)	Ratio of electro- static decay (%)	Surface resis- tivity ( $\Omega/\text{cm.}$ )	Anti- block- ing proper- ties (class)	Coeff- icient of fric- tion
5	Run No.								
10	16	0.3	ethylene- propylene copolymer	given	2.5	40	$6 \times 10^{12}$	1	0.25
	17	0.5	ethylene- propylene copolymer	not given	2.5	10	$5 \times 10^{13}$	1	0.25
			ethylene- propylene copolymer	given	2.8	80	$1 \times 10^{12}$	1	0.25
15	18	0.7	ethylene- propylene copolymer	not given	2.6	40	$6 \times 10^{12}$	1	0.25
			ethylene- propylene copolymer	given	2.9	85	$8 \times 10^{11}$	1	0.25
	19	1.0	ethylene- propylene copolymer	not given	2.6	40	$6 \times 10^{12}$	1	0.25
			ethylene- propylene copolymer	given	3.0	90	$3 \times 10^{11}$	1	0.25
20	20	0.5	low den- sity poly- ethylene	not given	2.8	45	$5 \times 10^{12}$	1	0.25
			ethylene	given	3.5	85	$8 \times 10^{11}$	2	0.40
	21	0.5	high den- sity poly- ethylene	not given	3.3	60	$2 \times 10^{12}$	1—2	0.40
25			ethylene	given	3.5	80	$1 \times 10^{12}$	1—2	0.27
	22	0.5	ethylene- butene copolymer	not given	3.3	55	$3 \times 10^{12}$	1	0.25
			ethylene- butene	given	2.5	40	$6 \times 10^{12}$	4	0.90
30	23	0.5	ethylene- propylene- butene terpolymer	not given	2.5	20	$1 \times 10^{13}$	4	0.90
			ethylene- propylene- butene	given	2.5	80	$1 \times 10^{12}$	1	0.25
			ethylene- propylene- butene	not given	2.5	40	$6 \times 10^{12}$	1	0.25
35	24	0.5	ethylene- butene- (40 parts)	given	3.2	50	$4 \times 10^{12}$	3	0.50
			high den- sity PE (60 parts)	not given	3.0	30	$7 \times 10^{12}$	2—3	0.50
40	25	0.5	same mix- ture as above	given	3.5	90	$3 \times 10^{11}$	1—2	0.25
			also contain- ing silica (0.2 part)	not given	3.5	50	$4 \times 10^{12}$	1	0.25
45			and oleic amide (0.5 part)						
	26	0.1	ethylene- propylene- copolymer	given	2.3	5	$1 \times 10^{14}$	1	0.25
			ethylene- propylene- copolymer	not given	2.3	0	$1 \times 10^{16}$	1	0.25
50	27	1.2	ethylene- propylene copolymer	given	4.0	100	or higher $2 \times 10^{11}$	1	0.25
			ethylene- propylene copolymer	not given	3.5	45	$5 \times 10^{12}$	1	0.25

## Example 4

A uniaxially stretched polypropylene film was prepared, from a composition comprising 100 parts by weight of the same polypropylene pellets used in Example 1 having incorporated therein 0.1 part by weight of stearic acid, and a mixture of palmityl monoglyceride and stearyl monoglyceride in the ratio of 30:70 added as an anti-static agent as in Example 1. Each of the ethylene-( $\alpha$ -olefin) copolymer resins listed in Table 4 was extrusion laminated on both surfaces of the respective sample of the uniaxially stretched polypropylene film prepared as noted above to provide lamination layers each 20 microns thick on the substrate forming an integral laminated film 200 microns thick. The resulting laminated film was then transversely stretched as in Example 1 to provide a biaxially oriented polypropylene composite film 25 microns thick in one continuous process.

The composite film thus prepared was aged in the atmosphere at a temperature of 40°C. for 48 hours before it was tested as in Example 1 to determine its characteristic properties, including anti-static properties, anti-blocking properties, transparency, lubricity and the like.

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The uniaxially oriented polypropylene composite film prepared as mentioned above was further treated by corona discharge on one surface thereof and then aged for 48 hours in the atmosphere at a temperature of 40°C. The characteristic properties of the resulting treated film were determined in the same manner as noted hereinbefore.

In the column under the heading "surfaces measured" in Table 4, the symbol A means that "the ratio of electrostatic decay" and "the surface resistivity" of the corresponding sample were measured with respect to the corona treated surface of the composite film; and the "anti-blocking properties" and the "coefficient of friction" were measured with respect to the contact of the two corona treated surfaces.

The symbol B means that "ratio of electrostatic decay" and "surface resistivity" were measured with respect to the non-corona treated surface; and "blocking properties" and "coefficient of friction" were measured with respect to the contact of the two non-corona discharge treated surfaces of the composite films.

The symbol C means that the "ratio of electrostatic decay" and "the surface resistivity" were measured with respect to the inner side surface of a rolled composite film, and both the "blocking properties" and the "coefficient of friction" were measured with respect to the two contacting inner side surfaces of a rolled composite film.

The symbol D means that "ratio of electrostatic decay" and "surface resistivity" were measured with respect to the outer surface of a rolled composite film; and "blocking properties" and "coefficient of friction" were measured with respect to the contacting surfaces placed in such a manner that the outer surfaces of two rolled composite films were contacting each other.

Table 4

Run No.	Type of polymer used as lamination layer	Corona discharge treatment	Haze (%)	Surfaces measured	Ratio of electrostatic decay (%)	Surface resistivity ( $\Omega/\text{cm.}$ )	Anti-blocking properties (class)	Coefficient of friction
28	ethylene-propylene copolymer	given	3.7	A	90	$3 \times 10^{11}$	1	0.25
30				B	90	$3 \times 10^{11}$	1	0.25
		not given	3.6	C	55	$3 \times 10^{12}$	1	0.25
				D	55	$3 \times 10^{12}$	1	0.25
29	low density polyethylene	given	4.5	A	90	$3 \times 10^{11}$	2	0.40
				B	90	$3 \times 10^{11}$	1—2	0.40
35		not given	4.3	C	60	$2 \times 10^{12}$	1—2	0.40
				D	60	$2 \times 10^{12}$	1—2	0.40
30	high density polyethylene	given	4.5	A	85	$8 \times 10^{11}$	1—2	0.27
				B	85	$8 \times 10^{11}$	1	0.25
		not given	4.3	C	60	$2 \times 10^{12}$	1	0.25
				D	60	$2 \times 10^{12}$	1	0.25
40	ethylene-butene copolymer	given	3.0	A	60	$2 \times 10^{12}$	4	0.90
				B	60	$2 \times 10^{12}$	4	0.90
		not given	3.0	C	30	$2 \times 10^{12}$	4	0.90
				D	30	$2 \times 10^{12}$	4	0.90
45	ethylene-propylene-butene terpolymer	given	3.7	A	90	$3 \times 10^{11}$	1	0.25
				B	90	$3 \times 10^{11}$	1	0.25
		not given	3.5	C	60	$2 \times 10^{12}$	1	0.25
				D	60	$2 \times 10^{12}$	1	0.25
33	a mixture of ethylene-butene (40 parts) and high density polyethylene (60 parts)	given	3.7	A	60	$2 \times 10^{12}$	3	0.50
				B	60	$2 \times 10^{12}$	2—3	0.50
50		not given	3.5	C	50	$4 \times 10^{12}$	2—3	0.50
				D	50	$4 \times 10^{12}$	2—3	0.50
55	34 a mixture of ethylene-butene copolymer (40 parts), high density polyethylene (60 parts), silica (0.2 part) and oleic amide (0.5 part)	given	3.9	A	90	$3 \times 10^{11}$	1—2	0.25
				B	90	$3 \times 10^{11}$	1	0.25
		not given	3.8	C	80	$1 \times 10^{12}$	1	0.25
				D	80	$1 \times 10^{12}$	1	0.25
60								



When the biaxially oriented polypropylene composite films having excellent transparency, anti-blocking properties and lubricity properties prepared by Run Nos. 4 through 7, Nos. 12 through 21, No. 23 and No. 25 were used to make bags on a high speed automatic bag-making machine, it was proved that they were suitable for machine fabrication and there were no problems caused by an electrostatic charge.

The composite films having excellent anti-blocking properties and lubricity properties prepared by the processes of Run Nos. 28 through 30, No. 32 and No. 34 were used for packaging using a high speed overwrapping machine. No difficulties were encountered caused by electrostatic charge.

The following comparative experiments were carried out to demonstrate that if a fatty acid monoglyceride is incorporated in polypropylene to obtain a biaxially stretched polypropylene film according to the conventional process, the resulting biaxially stretched film does not exhibit substantial anti-static properties.

#### Comparative Example

A polypropylene composition comprising 100 parts by weight of polypropylene having an  $[\eta]$  determined in tetralin at 135°C. of 1.8, an insoluble residue determined in n-hexane of 90% or higher having incorporated therein 0.1 part by weight of stearic amide added as a lubricant, and 0.5 part by weight of stearyl-di- $\beta$ -hydroxyethylbetain added as an anti-static agent; and another polypropylene composition which is the same as mentioned above except that instead of said anti-static agent, 0.5 part by weight of stearyl monoglyceride was added, were melt extruded at an extrusion temperature of 280°C. to prepare respective films. These respective films were stretched in the direction of the machine to 5 times their initial length at a stretching temperature of 140°C. followed by stretching to 8 times the initial width and concomitantly orienting in a transverse direction to obtain the sequentially, biaxially oriented film 25 microns thick.

The resulting biaxially oriented film was aged in the atmosphere at a temperature of 40°C. for 48 hours and then a sample was taken from each film. For each sample, the anti-static properties, blocking properties, transparency, lubricity, etc., were determined.

Other samples of these biaxially stretched films were corona discharge treated on one surface of each film and placed in the atmosphere at a temperature of 40°C. for 48 hours before sampling to take samples for determining the above-mentioned characteristic properties. The results of these experiments are summarized in Table 5.

Table 5

Run No.	Anti-static agent	Corona discharge treatment	Haze (%)	Ratio of electrostatic decay (%)	Surface resistivity ( $\Omega/\text{cm.}$ )	Anti blocking properties (class)	Coef- ficient of friction
35	betain-type amphoteric surface active agent	given	1.2	40	$6 \times 10^{12}$ $1 \times 10^{16}$	1—2 1—2	0.50 0.55
40		not given	1.2	0	or higher		
36	stearyl monoglyceride	given	1.2	1.5	$1 \times 10^{16}$ or higher	1—2	0.55
45		not given	1.2	0	$1 \times 10^{16}$ or higher	1—2	0.55

For the corona discharge treated films, the "ratio of electrostatic decay" and the "surface resistivity" were measured with respect to the corona treated surface; and the "anti-blocking properties" and the "coefficient of friction" were measured with respect to the contact of the two corona treated surfaces.

In these Examples, the determination of the characteristic properties of the product films such as anti-static properties, haze, coefficient of friction, which show the lubricity, blocking properties and the like, were carried out by the following methods.

#### 1. Measurement of Anti-Static Properties

##### 1-1. Measurement of the Ratio of Electrostatic Decay (%)

The film was left for 24 hours in an atmosphere having a temperature of 20°C. and 65% RH. After that, it was charged at 6 KV for 1 minute using "Static Honestmeter" made by Shishido Shokai Co., Ltd., and then discharged for 30 seconds, and then the ratio of electrostatic decay (%) was determined.

##### 1-2. Surface Resistivity

The film was left for 24 hours in an atmosphere having a temperature of 20°C. and 65% RH.

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Then, the surface resistivity thereof was measured by the "Super Insulation Tester SM-10" made by Toa Dempa Co., Ltd.

## 2. Measurement of Haze

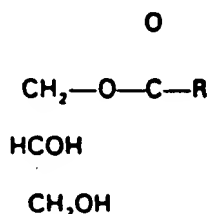
The haze was determined by the methods defined in JIS K-6714 and K-6718 using a haze meter available from Nihon Denshoku Kogyo Co., Ltd.

## 3. Measurement of Blocking Properties

Test pieces of film each 50 mm. x 50 mm. in size were prepared by random sampling and two of them were placed in contact with each other so that the surfaces to be tested were face to face. The two strips of film were left in an atmosphere of 40°C. and 90% RH for 24 hours under applied pressure of 2 Kgs. After that, the two strips of film were examined and evaluated in terms of the degree of blocking observed, the ranking ranging from Class 1 (no blocking) to Class 5 (Complete blocking).

## Claims

1. A process for the production of anti-static, biaxially oriented polypropylene composite films comprising forming a film from a polypropylene composition comprising 100 parts by weight of polypropylene and from about 0.3 to 1.0 part by weight of at least one fatty acid monoglyceride of the formula



- wherein R is a monovalent aliphatic hydrocarbon fatty acid residue;
- stretching said film in a first direction to obtain a uniaxially oriented film;
  - laminating polyethylene or an ethylene-( $\alpha$ -olefin) copolymer resin outer layer on at least one surface of said polypropylene film to form a laminated film; and
  - stretching the laminated film in the direction transverse to said first direction to form an anti-static, biaxially oriented polypropylene composite film.
  2. A process as claimed in claim 1, wherein said polypropylene film is formed by melt extrusion.
  3. A process as claimed in claim 1 or claim 2, wherein R is a saturated aliphatic hydrocarbon group having from 9 to 17 carbon atoms in the hydrocarbon group.
  4. A process as claimed in any one of claims 1 to 3, wherein said at least one outer layer is an ethylene-propylene copolymer layer.
  5. A process as claimed in any one of claims 1 to 3, wherein said at least one outer layer is a polyethylene layer.
  6. A process as claimed in any one of claims 1 to 3, wherein said at least one outer layer is an ethylene-butene copolymer layer.
  7. A process as claimed in any one of claims 1 to 3, wherein said at least one outer layer is an ethylene-propylene-butene terpolymer layer.
  8. A process as claimed in any one of claims 1 to 3, wherein said at least one outer layer is a mixture consisting of ethylene-butene copolymer and polyethylene.
  9. A process as claimed in any one of claims 1 to 8, wherein said polyethylene or ethylene-( $\alpha$ -olefin) copolymer resin contains an anti-blocking agent and a lubricant.
  10. A process as claimed in claim 9, wherein said anti-blocking agent is silica.
  11. A process as claimed in claim 9, wherein said lubricant is oleic amide.
  12. A process as claimed in any one of claims 1 to 11, wherein said fatty acid monoglyceride is decanoyl monoglyceride.
  13. A process as claimed in any one of claims 1 to 11, wherein said fatty acid monoglyceride is lauryl monoglyceride.
  14. A process as claimed in any one of claims 1 to 11, wherein said fatty acid monoglyceride is myristyl monoglyceride.
  15. A process as claimed in any one of claims 1 to 11, wherein said fatty acid monoglyceride is palmityl monoglyceride.
  16. A process as claimed in any one of claims 1 to 11, wherein said fatty acid monoglyceride is a mixture of palmityl monoglyceride and stearyl monoglyceride.
  17. An anti-static, biaxially oriented polypropylene composite film obtained by the process claimed in any one of claims 1 to 16.

18. A process as claimed in claim 1 and substantially as hereinbefore described with reference to any one of the Examples 1 to 4.

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